

## LA-UR-17-30696

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Title: Nuclear Forensics and Radiochemistry: Reaction Networks

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Intended for: Lecture Series at UC Berkeley

Issued: 2017-11-22

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# **Nuclear Forensics and Radiochemistry: Reaction Networks**

Robert S. Rundberg

## *Abstract:*

In the intense neutron flux of a nuclear explosion the production of isotopes may occur through successive neutron induced reactions. The pathway to these isotopes illustrates both the complexity of the problem and the need for high quality nuclear data. The growth and decay of radioactive isotopes can follow a similarly complex network. The Bateman equation will be described and modified to apply to the transmutation of isotopes in a high flux reactor. An alternative model of growth and decay, the GD code, that can be applied to fission products will also be described.

# Nuclear Forensics and Radiochemistry: Reaction Networks

## Lecture 5

# Reaction Networks

- A nuclear explosion produces an enormous flux of neutrons.
  - A neutron yield on the order of a mole ( $6.02E23$ ).
  - The chain reaction is finished in a short time, of the order of tens of nano-seconds.
  - The volume of burning fuel is relatively small.
- Multiple successive neutron induced reactions can occur.
- Activation of materials by successive reactions are prominent near the fuel.

# Neutron Reactions with Actinides (Uranium)

96	<b>Cm235</b> E 3.3	<b>Cm236</b> E 1.7 236.0514	<b>Cm237</b> E 2.7	<b>Cm238</b> 2.4 h E 1.0 238.05302	<b>Cm239(7/-)</b> ~3 h E 1.8	<b>Cm240</b> 27 d 240.055519	<b>Cm241 1/+</b> 32.8 d E .767	<b>Cm242</b> 162.8 d 242.058829	<b>Cm243 5/+</b> 29.1 a 243.061382	<b>Cm244</b> 18.1 a 244.062746
95	<b>Am234</b> 2.3 m E 4.2	<b>Am235</b> 15 m E 2.6	<b>Am236</b> 4 m E 3.3	<b>Am237 5/(-)</b> 1.22 h E 1.5	<b>Am238 1+</b> 1.63 h E 2.3	<b>Am239(5/-)</b> 11.9 h E .803	<b>Am240(3-)</b> 2.12 d E 1.38	<b>Am241 5/-</b> 432.7 a 241.056823	<b>Am242 1-</b> 141 a 16.02 h	<b>Am243 5</b> 7.37E3 a 243.061373
94	<b>Pu233</b> 20.9 m E 2.1	<b>Pu234</b> 8.8 h E .39	<b>Pu235(5/+)</b> 25.3 m E 1.81	<b>Pu236</b> 2.87 a E .124	<b>Pu237 7/-</b> 45.2 d E .124	<b>Pu238</b> 87.7 a 238.049553	<b>Pu239 1/+</b> 2.410E4 a 239.052157	<b>Pu240</b> 6.56E3 a 240.053807	<b>Pu241 5/+</b> 14.4 a E .0208	<b>Pu242</b> 3.75E5 a 242.058737
93	<b>Np232(4+)</b> 14.7 m E 2.8	<b>Np233(5)</b> 36.2 m E 1.0	<b>Np234</b> 4.41 d E 1.14	<b>Np235</b> 3.8 d E .124	<b>Np236</b> 2.03 a E .124	<b>Np237 5/+</b> 2.14E6 a 237.04667	<b>Np238 2+</b> 2.117 d E 1.292	<b>Np239 5/+</b> 2.355 d E .722	<b>Np240(5+)</b> 7.22 m E 2.20	<b>Np241 5</b> 13.9 m E 1.3
92	<b>U231(5/-)</b> 4.2 d E .382	<b>U232</b> 69.8 a 232.037146	<b>U233</b> 1.592E5 a 233.039628	<b>U234</b> 2.46E5 a 234.040952	<b>U235</b> 7.04E8 a 235.043924	<b>U236</b> 2.342E7 a 236.045563	<b>U237 1/+</b> 6.75 a 237.04667	<b>U238</b> 4.47E9 a 238.050783	<b>U239 5/+</b> 23.47 m E 1.263	<b>U240</b> 14.1 h E .39

$^{234}\text{U}(n,2n)^{233}\text{U}$   
Threshold 6.9 MeV eV  
0.4 barn @14 MeV eV

$^{235}\text{U}(n,\text{fission})^{235}\text{U}$   
1.3 barn @2 eV

$^{235}\text{U}(n,\gamma)^{236}\text{U}$   
3 barn @1 keV

$^{236}\text{U}(n,\gamma)^{237}\text{U}$   
3 barn @1 keV

# Neutron Reactions with Actinides (Uranium)

96	<b>Cm235</b> E 3.3	<b>Cm236</b> E 1.7 236.0514	<b>Cm237</b> E 2.7	<b>Cm238</b> 2.4 h E 1.0 238.05302	<b>Cm239(7/-)</b> ~3 h E 1.8	<b>Cm240</b> 27 d 240.055519	<b>Cm241 1/+</b> 32.8 d E .767	<b>Cm242</b> 162.8 d 242.058829	<b>Cm243 5/+</b> 29.1 a 243.061382	<b>Cm244</b> 18.1 a 244.062746	
95	<b>Am234</b> 2.3 m E 4.2	<b>Am235</b> 15 m E 2.6	<b>Am236</b> 4 m E 3.3	<b>Am237 5/(-)</b> 1.22 h E 1.5	<b>Am238 1+</b> 1.63 h E 2.3	<b>Am239(5/-)</b> 11.9 h E .803	<b>Am240(3-)</b> 2.12 d E 1.38	<b>Am241 5/-</b> 432.7 a 241.056823	<b>Am242 1-</b> 141 a 242.061373	<b>Am243 5</b> 7.37E3 a 243.061373	
94	<b>Pu233</b> 20.9 m E 2.1	<b>Pu234</b> 8.8 h E .39	<b>Pu235(5/+)</b> 25.3 m E 1.14	<b>Pu236</b> 2.87 a 236.046048	<b>Pu237 7/-</b> 45.2 d E 2.2	<b>Pu238</b> 87.7 a E 2.3	<b>Pu239 1/+</b> 2.410E4 a E 2.3	<b>Pu240</b> 6.56E3 a E 2.3	<b>Pu241 5/+</b> 14.4 a E .0208	<b>Pu242</b> 3.75E5 a 242.058737	
93	<b>Np232(4+)</b> 14.7 m E 2.8	<b>Np233(5/+)</b> 36.2 m E 1.0	<b>Np234(0+)</b> 4.4 d E 1.81	<b>Np235 5/+</b> 1.085 a E .124	<b>1(-)Np236</b> 22.5 h E 2.2	<b>1(-)Np237</b> 2.14E4 a E 2.3	<b>1(-)Np238</b> 2.14E4 a E 2.3	<b>1(-)Np239</b> 2.14E4 a E 2.3	<b>1(+ )Np240 (5+)</b> 2.2 m E 2.3	<b>Np241 5</b> 13.9 m E 2.3	
92	<b>U231 (5/-)</b> 4.2 d E .382	<b>U232</b> 69.8 a 232.037146	<b>U233 5/+</b> 1.592E5 a 233.039628	<b>U234</b> 0.0055 a 234.040946	<b>1/+ U235 7/-</b> 26 m 235.043923	<b>AcU</b> 0.7200 a 7.04E8 a	<b>U236</b> 2.342E7 a 236.045562	<b>U237 1/+</b> 6.75 d 237.04816	<b>U238</b> 99.2745 a 238.02891	<b>U239 5/+</b> 23.47 m 239.052163	<b>U240</b> 14.1 h 240.056581

$^{237}\text{U}(n,3n)^{236}\text{U}$   
 Threshold 11.3 MeV  
 0. 0.4 barn @14 MeV

$^{238}\text{U}(n,\gamma)^{239}\text{U}$      $^{239}\text{U}(n,\gamma)^{240}\text{U}$   
 0.4 barn @1 k    3.0 barn @1 keV

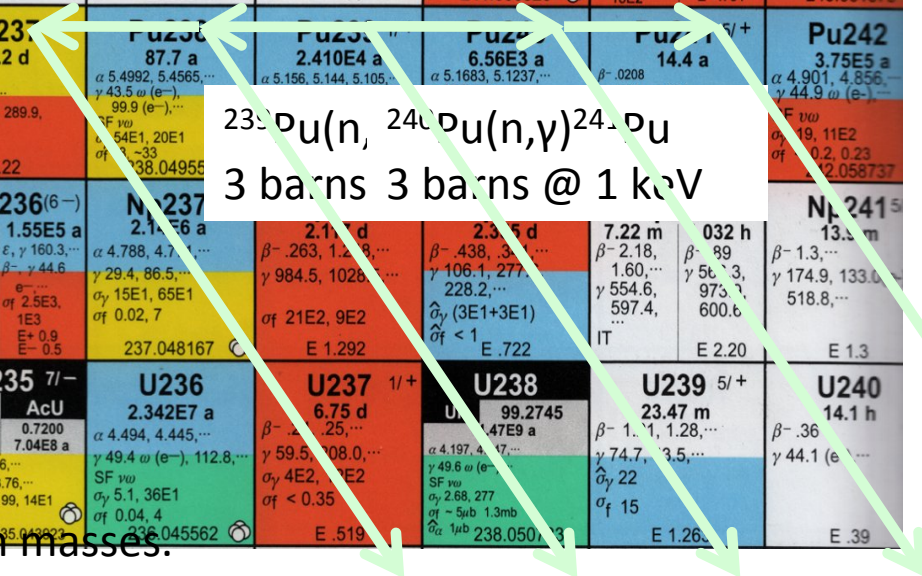
# Neutron Reactions with Actinides (Plutonium)

96	<b>Cm235</b> E 3.3	<b>Cm236</b> E 1.7 236.0514	<b>Cm237</b> E 2.7	<b>Cm238</b> 2.4 h E 1.0 238.05302	<b>Cm239(7/-)</b> ~3 h E 1.87	<b>Cm240</b> 27 d E 6.291, 6.248, ... E 44.6	<b>Cm241 1/+</b> 32.8 d E 4.718, ... E 5.939 (w),	<b>Cm242</b> 162.8 d E 6.1127, 6.0694, ... E 44.1 (e-), ...	<b>Cm243 5/+</b> 29.1 a E 5.785, 5.742, ... E 277.6, 228.2, ...	<b>Cm244</b> 18.1 a E 5.8048, 5.7627, ... E 42.8 (e-), ...	
95	<b>Am234</b> 2.3 m E 6.46 ?	<b>Am235</b> 15 m E ?	<b>Am236</b> 4 m E ?	<b>Am237</b> 1.22 h E 280.2, ... E 6.04 w	<b>Am238</b> E 561.0, ... E 5.94 w	<b>Am239</b> E 5.774 (w), 5.734, ... E 49.3 w	<b>Am240</b> E 5.378 w, ...	<b>Am241</b> E 5.156, 5.144, 5.105, ...	<b>Am242 1-</b> 141 a E 48.6, e- E 5.207 w, ...	<b>Am243 5/+</b> 7.37E3 a E 5.276, 5.234, ... E 74.7, 31.1, 662.2	
94	<b>Pu233</b> 20.9 m E 235.3, 534.7, ... E 6.30 w	<b>Pu234</b> 8.8 h E 6.200, 6.149, ...	<b>Pu235(5/+)</b> 25.3 m E 49.2, 756.4, ... E 5.85 w	<b>Pu236</b> 2.87 a E 5.7675, 5.7209, ... E 47.6-643.7 w E 236.046048	<b>Pu237</b> 45.2 d E 59.5, ... E 5.344(w), ... E 280.4(w), 289.9, ... E 320.8, ... E 54E1, 20E1 E 338.04955	<b>Pu238</b> 87.7 a E 5.4992, 5.4565, ... E 43.5 w (e-), ... E 99.9 (e-), ... E 54E1, 20E1 E 338.04955	<b>Pu239</b> 2.410E4 a E 5.156, 5.144, 5.105, ...	<b>Pu240</b> 6.56E3 a E 5.1683, 5.1237, ...	<b>Pu241 5/+</b> 14.4 a E 4.901, 4.856, ... E 44.9 w (e-), ...	<b>Pu242</b> 3.75E5 a E 4.901, 4.856, ... E 44.9 w (e-), ...	
93	<b>Np232(4+)</b> 14.7 m E 326.8, 819.2, ... E 866.8, 864.3, ...	<b>Np233(5/+)</b> 36.2 m E 312.0, 298.9, ... E 546.5, ... E 5.53 ? w	<b>Np234(0+)</b> 4.4 d E 79 w, ... E 1558.7, 1527.2, ... E 1602.2, ... E 9E2	<b>Np235 5/+</b> 1.085 a E 5.021 (w), ... E 5.004, ... E 25.6-188.8 w E 15E1+?	<b>Np236(6-)</b> 22.5 h E 59.5, ... E 160.3, ... E 54, ... E 642.3, ... E 687.6, ... E 2.7E3, ... E 7E2	<b>Np237</b> 2.14E6 a E 4.788, 4.7, ... E 29.4, 86.5, ... E 15E1, 65E1 E 0.02, 7	<b>Np238</b> 2.11E5 d E 2.1, ... E 263, 1.2, ... E 984.5, 1026, ... E 21E2, 9E2	<b>Np239</b> 2.35E5 d E 438, 3, ... E 106.1, 277, ... E 228.2, ... E 3E1+3E1 E 722	<b>Np240</b> 7.22 m E 2.18, ... E 1.60, ... E 554.6, ... E 597.4, ... E 600.6	<b>Np241 5/+</b> 0.32 h E 89, ... E 56.3, ... E 973, ... E 600.6	<b>Np242 5/+</b> 13.2 m E 1.3, ... E 174.9, 133.0, ... E 518.8, ...
92	<b>U231(5/-)</b> 4.2 d E 25.65, 84.2, ... E 5.456 w, ... E 68.33, 53.23, ... E 3E2	<b>U232</b> 69.8 a E 5.3203, 5.2635, ... E 57.8 w (e-), ... E 129.1, ... E 46.11E1 E 73, 28E1 E 75, 38E1	<b>U233 5/+</b> 1.592E5 a E 4.824, 4.783, ... E 42.5 w, 97.1, 54.7, ... E 46.11E1 E 73, 28E1 E 75, 38E1	<b>U234</b> 0.0055 a E 4.776, 4.725, ... E 53.2 (e-), 120.9, ... E 185.72, 143.76, ... E 585, -275	<b>U235 7/-</b> 26 m E 76.8 eV E 0.7200 E 7.04E8 a	<b>U236</b> 2.342E7 a E 4.494, 4.445, ... E 49.4 w (e-), 112.8, ... E 5.1, 36E1 E 0.04, 4	<b>U237 1/+</b> 6.75 d E 2.25, ... E 59.5, 108.0, ... E 4E2, 1E2 E < 0.35	<b>U238</b> 99.2745 a E 4.197, 4.17, ... E 49.6 w (e-), ... E 2.68, 277 E 5 w, 1.3 mb E 238.0501, ...	<b>U239 5/+</b> 23.47 m E 1.1, 1.28, ... E 74.7, 3.5, ... E 22 E 15	<b>U240</b> 14.1 h E 36, ... E 44.1 (e-), ...	

$^{238}\text{Pu}(n,2n)^{237}\text{Pu}$   
Threshold 7.0 MeV  
0.12 barn @ 14 MeV

$^{239}\text{Pu}(n, \gamma)^{240}\text{Pu}$   
3 barns @ 1 keV

Depletion by fission,  
Threshold fission for even masses.





# Neutron Reactions with the Actinides (Americium)

96	<b>Cm235</b> E 3.3	<b>Cm236</b> E 1.7 236.0514	<b>Cm237</b> E 2.7	<b>Cm238</b> 2.4 h E 1.0 238.05302	<b>Cm239</b> E 1.8	<b>Cm240</b> E .767	<b>Cm241</b> E 242.058829	<b>Cm242</b> E 243.062746	<b>Cm244</b> 18.1 a α 5.8048, 5.7627, ... γ 42.8 ω (e-), ... SF νω σ <sub>γ</sub> 15, -65E1 σ <sub>f</sub> 1.0, 11			
	<b>Am234</b> 2.3 m α 6.46 ?	<b>Am235</b> 15 m ε ?	<b>Am236</b> 4 m ε	<b>Am237</b> <sup>5/(-)</sup> 1.22 h ε γ 280.2, ... α 6.04 ω	<b>Am238</b> <sup>1+</sup> 1.63 h ε β <sup>+</sup> , ω γ 962.8, 946.7, ... 561.0, ... α 5.94 νω	<b>Am239</b> <sup>5/(-)</sup> 11.9 h ε γ 277.6, 228.2, ...	<b>Am240</b> <sup>5/(-)</sup> 2.12 d ε γ 987.7, 888.8, ...	<b>Am241</b> <sup>5/(-)</sup> 432.7 a α 5.4857, 5.4430, ... γ 59.5, 26.3-955	<b>Am242</b> <sup>5/(-)</sup> 141 a IT 48.6, e- γ 5.207ω, ... σ <sub>γ</sub> 2.2ω, ... SF νω σ <sub>γ</sub> 2E2, ... σ <sub>f</sub> 70E2, 18E2	<b>Am243</b> <sup>5/(-)</sup> 7.37E3 a α 5.276, 5.234, ... γ 74.7, 31.1-662.2 SF νω σ <sub>γ</sub> (74+), ... E -0.074, 0.06 243.061373		
95	<b>Pu233</b> 20.9 m ε γ 235.3, 534.7, ... α 6.30 ω	<b>Pu234</b> 8.8 h ε α 6.200, 6.149, ...	<b>Pu235</b> <sup>5/+</sup> 25.3 m ε γ 49.2, 756.4, ... α 5.85 ω	<b>Pu236</b> 2.87 a α 5.7675, 5.7209, ... γ 47.6-643.7 ω SF νω σ <sub>f</sub> 16E1, 1E3	<b>Pu237</b> 45.1 m ε, γ 59.5, ... α 5.344(ω), ... γ 280.4(νω), 289.9, ... 320.8, ... σ <sub>f</sub> 24E2	<b>Pu238</b> E .22 α 5.4992, 5.4565, ... γ 43.5 ω (e-), ... 99.9 (e-), ... SF νω σ <sub>γ</sub> 54E1, 20E1 σ <sub>f</sub> 18, -33	<b>Pu239</b> 238.049553 α 5.156, 5.144, 5.105, ... γ 51.6 e-, 30.1-1057.3ω SF νω σ <sub>γ</sub> 271, 20E1 σ <sub>γ</sub> 750, 30E1 σ <sub>α</sub> < 0.4 mb	<b>Pu240</b> 56E3 a α 5.1683, 5.1237, ... γ 45.2 ω (e-), ... 104.2(e-), ... SF νω σ <sub>γ</sub> 290, 81E2 σ <sub>f</sub> 0.05, 2.4	<b>Pu241</b> <sup>5/+</sup> 14.4 a β- 0208 α 4.897 ω, 4.853, ... γ 148.6 (νω), 103.7, ... σ <sub>γ</sub> -361, 16E1 σ <sub>γ</sub> 101E1, 57E1 σ <sub>α</sub> < 0.2 mb	<b>Pu242</b> 3.75E5 a α 4.901, 4.856, ... γ 44.9 ω (e-), ... SF νω σ <sub>γ</sub> 19, 11E2 σ <sub>f</sub> < 0.2, 0.23 242.058737		
	<b>94</b>	<b>93</b>	<b>92</b>	<b>91</b>	<b>90</b>	<b>89</b>	<b>88</b>	<b>87</b>	<b>86</b>	<b>85</b>		
<b>93</b>	<b>Np232</b> <sup>4+</sup> 14.7 m ε γ 326.8, 819.2, ... 866.8, 864.3, ...	<b>Np233</b> <sup>5/+</sup> 36.2 m ε γ 312.0, 298.9, ... 546.5, ... α 5.53 ? ω	<b>Np234</b> <sup>0+</sup> 4.4 d ε β <sup>+</sup> .79 ω γ 1558.7, 1527.2, ... 1602.2, ... σ <sub>f</sub> 9E2	<b>Np235</b> <sup>5/+</sup> 1.085 a ε α 5.021 (νω), ... 5.004, ... γ 25.6-188.8 νω σ <sub>γ</sub> (15E1+?)	<b>1(-)Np236</b> <sup>6-</sup> 22.5 h ε, β- γ 54, ... σ <sub>f</sub> 2.7E3, 7E2	<b>1.55E5 a</b> ε, γ 160.3, ... β- γ 44.6 σ <sub>f</sub> 2.5E3, 1E3 E- 0.9 E- 0.5	<b>Np237</b> <sup>5/+</sup> 2.14E6 a α 4.788, 4.771, ... γ 29.4, 86.5, ... σ <sub>γ</sub> 15E1, 65E1 σ <sub>f</sub> 0.02, 7	<b>Np238</b> <sup>2+</sup> 2.117 d β- 263, 1.248, ... γ 984.5, 1028.5, ... σ <sub>f</sub> 21E2, 9E2	<b>Np239</b> <sup>5/+</sup> 2.355 d β- 438, 341, ... γ 106.1, 277.6, ... 228.2, ... σ <sub>γ</sub> (3E1+3E1) σ <sub>f</sub> < 1	<b>1(+ )Np240</b> <sup>5+</sup> 7.22 m β- 2.18, ... 1.60, ... γ 554.6, ... 597.4, ... IT	<b>1.032 h</b> β- .89 γ 566.3, ... 973.9, ... 600.6	<b>Np241</b> <sup>5</sup> 13.9 m β- 1.3, ... γ 174.9, 133.0(e-), ... 518.8, ...
<b>92</b>	<b>U231</b> <sup>5/(-)</sup> 4.2 d ε γ 25.65, 84.2, ... α 5.456 ω, ... γ 68.33, 53.23, ... σ <sub>f</sub> 3E2	<b>U232</b> 69.8 a α 5.3203, 5.2635, ... γ 57.8 ω (e-), ... 129.1, ... SF ω σ <sub>γ</sub> 73, 28E1 σ <sub>f</sub> 75, 38E1	<b>U233</b> <sup>5/+</sup> 1.592E5 a α 4.824, 4.783, ... γ 42.5 ω, 97.1, 54.7, ... SF νω σ <sub>γ</sub> 46, 14E1 σ <sub>γ</sub> 531, 76E1 σ <sub>α</sub> < 0.3 mb	<b>U234</b> 0.0055 2.46E5 a α 4.776, 4.725, ... γ 53.2 (e-), 120.9, ... SF νω σ <sub>γ</sub> 100, 7E2 σ <sub>f</sub> < 5 mb, 7	<b>1/+ U235</b> <sup>7/(-)</sup> 26 m IT -76.8 eV (ω), e- 0.7200 7.04E8 a α 4.398, 4.366, ... γ 185.72, 143.76, ... SF νω σ <sub>γ</sub> 585, -275 σ <sub>α</sub> < .1 mb 235.043923	<b>U236</b> 2.342E7 a α 4.494, 4.445, ... γ 49.4 ω (e-), 112.8, ... SF νω σ <sub>γ</sub> 5.1, 36E1 σ <sub>f</sub> 0.04, 4	<b>U237</b> <sup>1/+</sup> 6.75 d β- 24, 25, ... γ 59.5, 208.0, ... σ <sub>γ</sub> 4E2, 12E2 σ <sub>f</sub> < 0.35	<b>U238</b> 99.2745 4.47E9 a α 4.197, 4.147, ... γ 49.6 ω (e-), ... SF νω σ <sub>γ</sub> 2.68, 277 σ <sub>f</sub> -5mb 1.3mb σ <sub>α</sub> 1 μb 238.050783	<b>U239</b> <sup>5/+</sup> 23.47 m β- 1.21, 1.28, ... γ 74.7, 43.5, ... σ <sub>γ</sub> 22 σ <sub>f</sub> 15	<b>U240</b> 14.1 h β- .36 γ 44.1 (e-), ...		

<sup>241</sup>Am beta decay to <sup>242</sup>Cm @ 1 keV @ 1 MeV

0.3 percent <sup>241</sup>Pu Beta decay to <sup>241</sup>AM

# Actinide Isotopes in Nuclear Debris

- The equations for the successive reactions are solved numerically in a code using the Runge-Kutta method.
- The isotopes produced give the neutron fluence and spectrum that the original material was exposed to.

# The Advantage of Isotope Ratios

If we assume no burnup the first order equation is

$$\frac{dN_{A-1}}{dt} = \sigma_1 N_A \phi. \quad (1)$$

Assume constant flux, the solution is

$$N_{A-1} = \sigma_1 N_A \phi t. \quad (2)$$

The second order reaction is then

$$\frac{dN_{A-2}}{dt} = \sigma_2 [\sigma_1 N_A \phi t] \phi. \quad (3)$$

The solution is

$$\begin{aligned} N_{A-2} &= \sigma_1 \sigma_2 N_A \int_0^t \phi^2 t dt \\ &= \frac{\sigma_1 \sigma_2 N_A (\phi t)^2}{2}. \end{aligned} \quad (4)$$

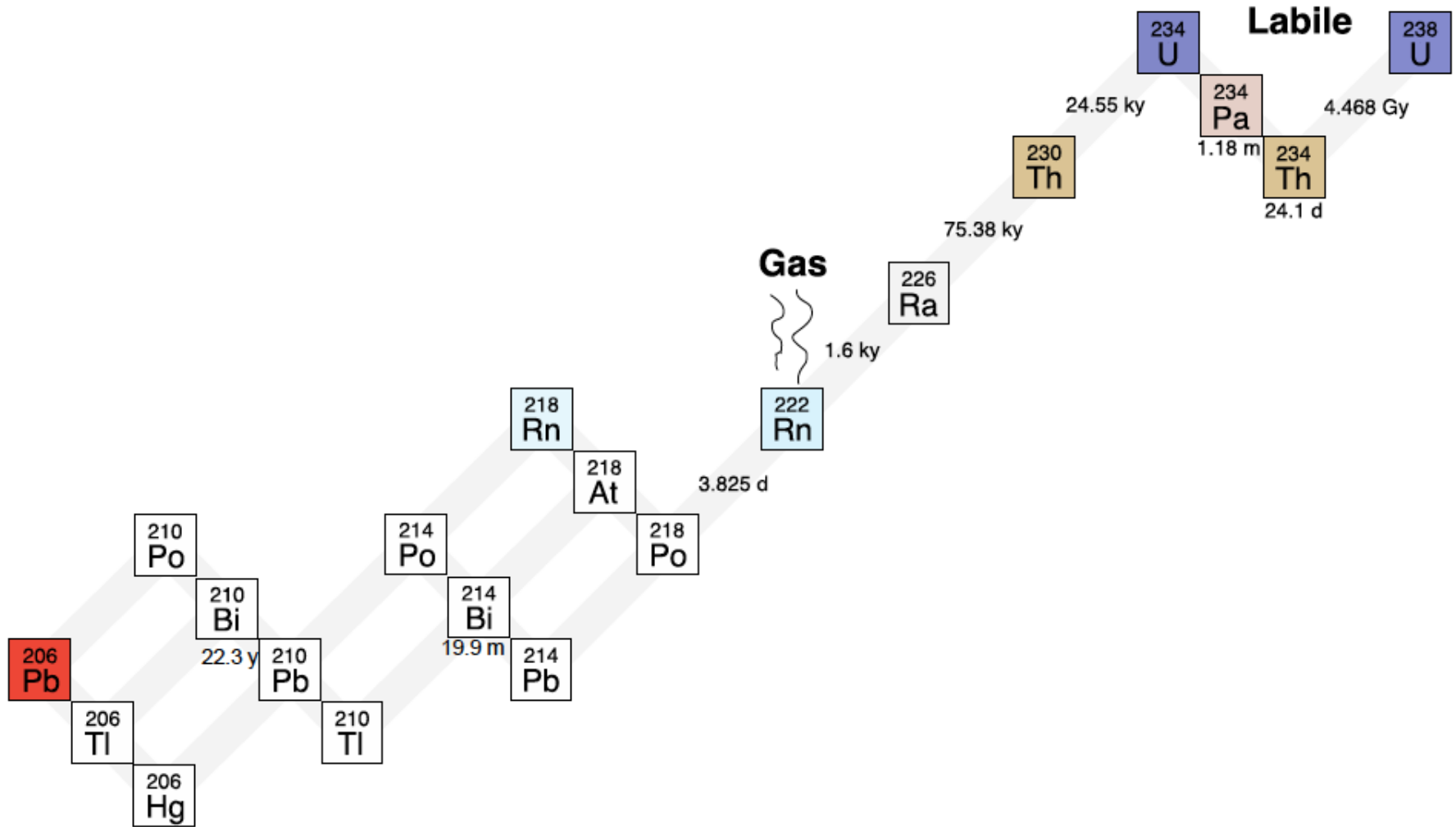
Therefore the atom ratio is

$$\frac{N_{A-2}}{N_{A-1}} = \frac{\sigma_2}{2} \phi t. \quad (5)$$

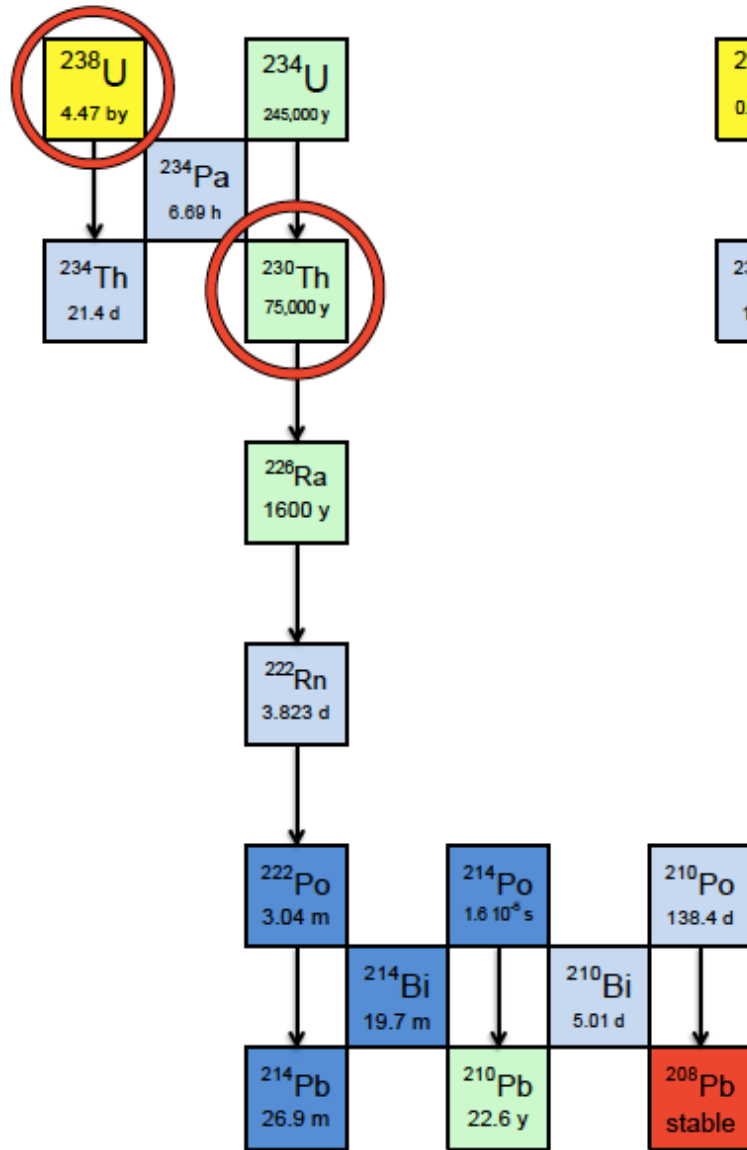
# Growth and Decay

- The growth and decay of radioactive isotopes are similar to the transmutation of isotopes in high flux environment.
- The Bateman equations are useful. But restricted to specific initial conditions.
- The Bateman equations can be modified to include transmutation.

# $^{238}\text{U}$ Natural Decay Series



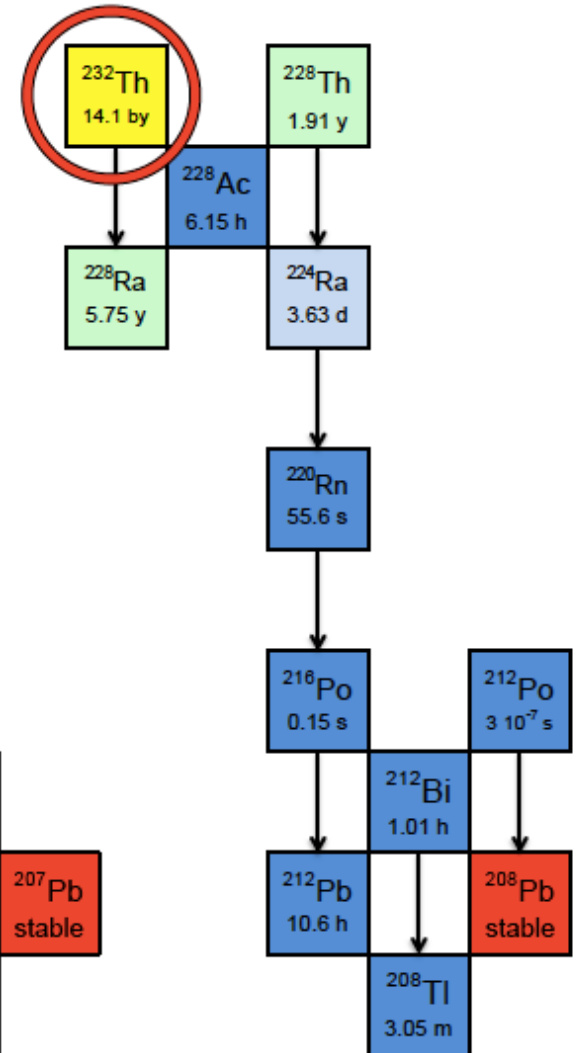
### $^{238}\text{U}$ -series



### $^{235}\text{U}$ -series



### $^{232}\text{Th}$ -series



# Bateman Equations

Start with a simple parent/daughter growth and decay. The equation for the daughter is,

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

or

$$\frac{dN_2}{dt} + \lambda_2 N_2 - \lambda_1 N_1^0 e^{-\lambda_1 t} = 0. \quad (1)$$

The solution of this linear differential equation of the first order may be obtained by standard methods and gives

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2^0 e^{-\lambda_2 t}. \quad (2)$$

# Bateman Equations

Consider the granddaughter. The equation for its growth and decay is,

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3. \quad (3)$$

Eq. 3 is analogous to Eq. 1, but the solution calls for more labor, because  $N_2$  is a much more complicated function than  $N_1$ . The great granddaughter is even more complicated. Fortunately, H. Bateman has given the solution for a chain of  $n$  members with the special assumption that at  $t = 0$  the parent substance alone is present, that is,  $N_2^0 = N_3^0 = \dots = N_n^0 = 0$ . This solution is

$$\begin{aligned} N_n &= C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \dots + C_n e^{-\lambda_n t}, \\ C_1 &= \frac{\lambda_1 \lambda_2 \dots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1) \dots (\lambda_n - \lambda_1)} N_1^0, \\ C_2 &= \frac{\lambda_1 \lambda_2 \dots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2) \dots (\lambda_n - \lambda_2)} N_1^0, \text{ and so on.} \end{aligned} \quad (4)$$



# Bateman Applied to Transmutation

Successive neutron reactions in a high flux, such as, a high flux reactor can be solved using the Bateman equations, as well. The rate of disappearance of an isotope in a neutron flux is

$$-\frac{dN}{dt} = (\lambda + n\nu\sigma)N = \Lambda N. \quad (5)$$

Consider a parent daughter pair. The parent disappears by both transmutation and decay. But the daughter grows by decay of the parent only and disappears by both processes. In general notation,

$$\frac{dN_{i+1}}{dt} = \lambda_i N_i - \Lambda_{i+1} N_{i+1}.$$

We replace  $\lambda_i$  by a modified decay constant  $\Lambda_i^* = \lambda_i^* + n\nu\sigma_i^*$ , were only the decay constant of transmutation term that lead to next progeny in the chain is used.

# Bateman Applied to Transmutation

With this nomenclature the Bateman equation becomes,

$$N_n = C_1 e^{-\Lambda_1 t} + C_2 e^{-\Lambda_2 t} + \dots + C_n e^{-\Lambda_n t},$$

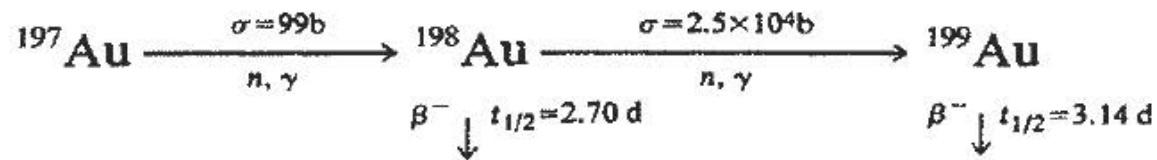
$$C_1 = \frac{\Lambda_1^* \Lambda_2^* \dots \Lambda_{n-1}^*}{(\Lambda_2 - \Lambda_1)(\Lambda_3 - \Lambda_1) \dots (\Lambda_n - \Lambda_1)} N_1^0,$$

$$C_2 = \frac{\Lambda_1^* \Lambda_2^* \dots \Lambda_{n-1}^*}{(\Lambda_1 - \Lambda_2)(\Lambda_3 - \Lambda_2) \dots (\Lambda_n - \Lambda_2)} N_1^0, \text{ and so on.}$$

(6)

# An Example

As an illustration, we compute the amount of 3.15-d  $^{199}\text{Au}$  formed by two successive  $(n, \gamma)$  reactions when 1 g  $^{197}\text{Au}$  is exposed for 30 h in a neutron flux of  $1 \times 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ . The chain of reactions is



We use (5-12) for this three-membered chain:

$$\begin{aligned}
 N_{199} = & \Lambda_{197}^* \Lambda_{198}^* N_{197}^0 \left[ \frac{e^{-\Lambda_{197}t}}{(\Lambda_{198} - \Lambda_{197})(\Lambda_{199} - \Lambda_{197})} \right. \\
 & \left. + \frac{e^{-\Lambda_{198}t}}{(\Lambda_{197} - \Lambda_{198})(\Lambda_{199} - \Lambda_{198})} + \frac{e^{-\Lambda_{199}t}}{(\Lambda_{197} - \Lambda_{199})(\Lambda_{198} - \Lambda_{199})} \right].
 \end{aligned}$$

The numerical values to be substituted are

$$t = 1.08 \times 10^5 \text{ s},$$

$$nv = 10^{14} \text{ cm}^{-2} \text{ s}^{-1},$$

$$\sigma_{197} = 9.9 \times 10^{-23} \text{ cm}^2,$$

$$\sigma_{198} = 2.5 \times 10^{-20} \text{ cm}^2,$$

$$N_{197}^0 = \frac{6.02 \times 10^{23}}{197} = 3.05 \times 10^{21},$$

$$\Lambda_{197}^* = \Lambda_{197} = nv\sigma_{197} = 9.9 \times 10^{-9} \text{ s}^{-1},$$

$$\begin{aligned} \Lambda_{198} &= \lambda_{198} + nv\sigma_{198} = 3.0 \times 10^{-6} + 2.5 \times 10^{-6} \\ &= 5.5 \times 10^{-6} \text{ s}^{-1}, \end{aligned}$$

$$\Lambda_{198}^* = nv\sigma_{198} = 2.5 \times 10^{-6} \text{ s}^{-1},$$

and

$$\Lambda_{199} = \lambda_{199} = 2.55 \times 10^{-6} \text{ s}^{-1}.$$

Using these values, we get

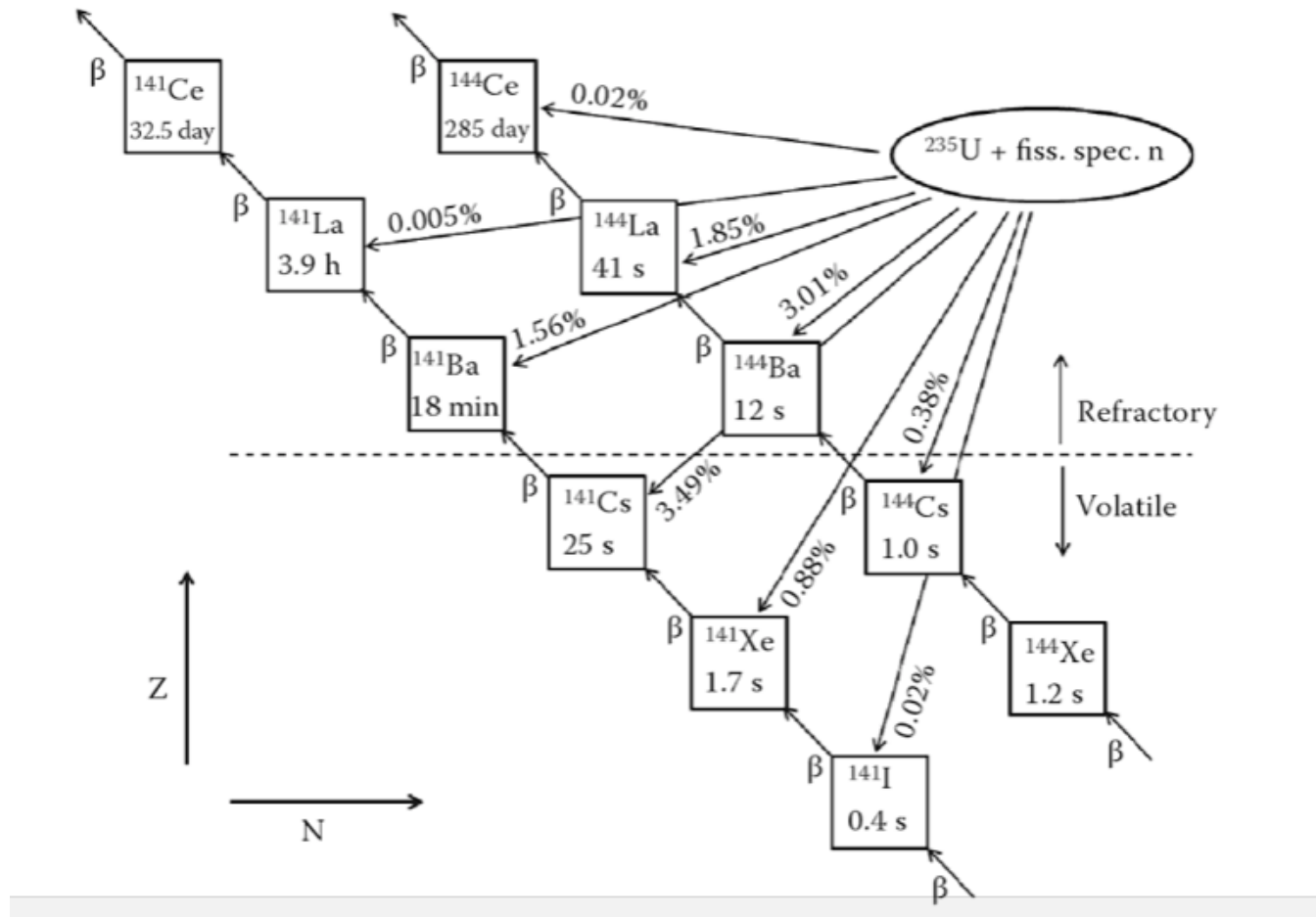
$$\begin{aligned}
 N_{199} &= 7.85 \times 10^7 \left( \frac{e^{-0.00107}}{5.5 \times 10^{-6} \times 2.55 \times 10^{-6}} \right. \\
 &\quad \left. + \frac{e^{-0.594}}{5.5 \times 10^{-6} \times 2.95 \times 10^{-6}} - \frac{e^{-0.275}}{2.55 \times 10^{-6} \times 2.95 \times 10^{-6}} \right) \\
 &= 7.55 \times 10^7 (7.12 \times 10^{10} + 3.40 \times 10^{10} - 1.01 \times 10^{11}) = 3.2 \times 10^{17}.
 \end{aligned}$$

The disintegration rate of  $^{199}\text{Au}$  at the end of the irradiation is  $\lambda_{199}N_{199} = 0.82 \times 10^{12} \text{ s}^{-1}$ . For comparison we compute the disintegration rate of  $^{198}\text{Au}$  in the sample [again from (5-12) for a two-membered chain]:

$$\begin{aligned}
 \lambda_{198}N_{198} &= \lambda_{198}n\nu\sigma_{197}N_{197}^0 \left( \frac{e^{-\Lambda_{197}t}}{\Lambda_{198} - \Lambda_{197}} + \frac{e^{-\Lambda_{198}t}}{\Lambda_{197} - \Lambda_{198}} \right) \\
 &= 9.06 \times 10^7 \frac{0.999 - 0.552}{5.5 \times 10^{-6}} = 7.36 \times 10^{12} \text{ s}^{-1}.
 \end{aligned}$$

Thus about 10 percent of the radioactive disintegrations in the sample occur in  $^{199}\text{Au}$ .

# Fission Does Not Always Meet the Initial Conditions



# The GD Code

The general differential equations for radioactive decay and growth

$$\begin{aligned}\frac{dN_1}{dt} &= a_{11}N_1 && +r_1(t) \\ \frac{dN_2}{dt} &= a_{21}N_1 + a_{22}N_2 && +r_2(t) \\ &\vdots && \\ \frac{dN_n}{dt} &= a_{n1}N_1 + \cdots + a_{nn}N_n && +r_n(t)\end{aligned}\tag{1}$$

where  $N_i(t)$  is the number of atoms of nuclide  $i$  existing at time  $t$ ,  $a_{ij}$ ,  $i \geq j$  are constants, and  $r_i$ 's are the rates of formation from sources other than by decay of isotopes  $i$ . A specialized form  $r_i(t) = y_i f(t)$  is used in the Los Alamos code GD, so that the rates have time independent ratios to each other. The  $y_i$ 's are called fractional independent yields.

# GD Code Continued

where  $\mathbf{A}$  is the matrix of  $a_{ij}$ 's,  $\mathbf{N}$  and  $\mathbf{Y}$  are column vectors and  $f(t)$  is a scalar. Equation (2) has the solution

$$\mathbf{N}(t) = e^{(t-\tau)\mathbf{A}}\mathbf{N}(\tau) + \int_{\tau}^t e^{(t-s)\mathbf{A}} f(s) \mathbf{Y} ds \quad (3)$$

There is a non singular matrix  $\mathbf{P}$  such that

$$\mathbf{P}^{-1}\mathbf{A}\mathbf{P} = \mathbf{J} \quad (4)$$

is diagonal. The GD code finds  $\mathbf{P}$ . The solution is then

$$\begin{aligned} \mathbf{N}(t) &= \mathbf{P} \operatorname{diag} \left[ e^{a_{11}(t-\tau)}, \dots, e^{a_{nn}(t-\tau)} \right] \mathbf{P}^{-1} \mathbf{N}(\tau) \\ &+ \mathbf{P} \left( \int_{\tau}^t \operatorname{diag} \left[ e^{a_{11}(t-s)}, \dots, e^{a_{nn}(t-s)} \right] f(s) ds \right) \mathbf{P}^{-1} \mathbf{Y} \end{aligned} \quad (5)$$



# The GD Code Solution

Define  $F_i(t)$  by

$$F_i(t - \tau) = \int_{\tau}^t e^{a_{ii}(t-s)} f(s) ds \quad (6)$$

then the solution is

$$\begin{aligned} \mathbf{N}(t) &= \mathbf{P} \operatorname{diag} \left[ e^{a_{11}(t-\tau)}, \dots, e^{a_{nn}(t-\tau)} \right] \mathbf{P}^{-1} \mathbf{N}(\tau) \\ &+ \mathbf{P} \operatorname{diag} [F_1(t - \tau), \dots, F_n(t - \tau)] \mathbf{P}^{-1} \mathbf{Y} \end{aligned} \quad (7)$$

The function  $f(s)$  is taken to be piecewise constant with  $f_j$  on time intervals  $U_j$  to  $W_j$ . With this specialization it follows that

$$F_i(t - \tau) = \frac{e^{a_{ii}t}}{-a_{ii}} \sum_j f_j (e^{-a_{ii}W_j} - e^{-a_{ii}U_j}). \quad (8)$$